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Applicants' respectfully disagree with the Examiner's position. A person of ordinary skill in the art who is reading Claim 1 in light of the supporting disclosure as is appropriate in a determination under Section 112, ¶2[1], finds on page 10, indicated line 21 et seq., of the application:

> *... a part TR of the remaining liquid phase is recirculated to the reactor and a part TA is discharged, ...*

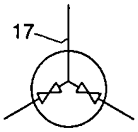
The first paragraph on page 16 of the application correspondingly provides:

> *... This solution is divided into a part TR which is recirculated straight back to the reactor cascade and a part TA which is discharged from the circuit. ...*

Additionally, applicants illustrate the division of the solution in a figure illustrating the invention, and state, in relation to that figure, on page 23 indicated line 20, of the application:

> *... an adjustable stream divider (18) for dividing the methanol stream [(17)] containing residual catalyst and catalyst degradation products leaving the distillation apparatus into the substreams TR and TA, ...*

The respective part of applicants' figure referenced in that statement of the application is reproduced below:



A person of ordinary skill in the art who is reading applicants' claims in light of the supporting disclosure is, therefore, immediately apprised that the requisite stream of the remaining liquid phase is split or divided into two substreams, one of which is recirculated to the reactor cascade and the other one of which is discharged. The designation of the recirculated part of the requisite liquid phase as *"TR"* and of the discharged part of the stream as *"TA"* can in no way be considered to confuse a person of ordinary skill to an extent where the subject matter of applicants' claims is rendered indefinite within the meaning of Section 112, ¶2. *"In rejecting a claim under the second paragraph of 35 U.S.C. 112, it is incumbent on the examiner to* *__establish__* *that one of ordinary skill in the pertinent art, when reading the claims in light of the supporting specification, would not have been able to ascertain with reasonable degree of*

---

1) Cf. Ex parte Wu, 10 USPQ2d 2031 at 2033 (BPAI 1989).

*precision and particularity the particular area set out and circumscribed by the claims"*2). The Examiner's statement that *"the abbreviations TR and TA in claim 1 are confusing because it is unclear what these abbreviations are referring to"* clearly fails to provide and argument which *establishes* that one of skill in the art is unable to ascertain the particular area set out and circumscribed in the claim when reading the claim in light of the disclosure. In light of the foregoing it is deemed unnecessary that applicants insert an additional explanation of the meaning of TR and TA in Claim 1 as requested by the Examiner.

Withdrawal of the rejection of Claims 1 to 4 under Section 112, ¶2, is respectfully solicited.

The Examiner has rejected Claims 1 to 4 under 35 U.S.C. §103(a) as being unpatentable in light of the disclosure of ***Chang et al.*** **(US 4,661,621)**.

In this context, the Examiner summarizes applicants' invention enumerating a part of the features which characterize applicants' process. A number of critical requirements of applicants' process are, however, not mentioned in the Examiner's summary. As clearly provided for in Claim 1, applicants' process for preparing methyl formate requires

1) that the reaction is carried out in a cascade comprising at least two reactor elements;
2) that the reaction is carried out at from 80 to 120°C;
3) that the reaction is carried out under a carbon monoxide pressure of from 90 to 180 bar;
4) that the reaction is carried out in the presence of from 0.05 to 0.5% by weight, based on the weight of the liquid feed, of an alkali metal alkoxide or alkaline earth metal alkoxide as catalyst;
5) that the molar ratio of carbon monoxide to methanol is set in the reaction at from 3:1 to 0.5:1;
6) that the total output from the reactor cascade is passed to a distilling apparatus in which essentially the methyl formate is stripped from the reaction mixture;
7) that the remaining liquid phase is split into a part TR and a part TA;

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2) See ftn. 1), page 2 of this paper, emphasis added.

8) that the part TR is from 20 to 80% of the remaining liquid phase;

9) that the part TR is recirculated to the reactor cascade;

10) that the part TA of the remaining liquid phase is discharged;

11) that residual catalyst and catalyst degradation products are removed solids-free from the discharged part TA in a desalting apparatus and the remaining methanol is returned directly or indirectly to the reactor.

As concerns requirement (1) of applicants' process the Examiner takes the position that ***Chang et al.***'s disclosure, and particular the illustration in Figure 1 thereof, provides for a cascade arrangement. Applicants respectfully disagree. In accordance with Figure 1 of ***Chang et al.*** the reaction is conducted in reaction zones or reactors (12) and (18). Reactor (12) is charged at the lower portion with a stream (10) of *"syngas"* and at the top portion with a stream of recycled methanol. Methanol is then, together with the product, withdrawn from (12) at the bottom and conveyed as stream (28) to the distillation zone (22). A stream (16) of unreacted CO³⁾ is withdrawn at the top of (12) and introduced as a bottom feed into reactor (18) which is charged in the upper part with recycled methanol. The reaction mixture resulting in reactor (18) is withdrawn in the bottom region and, separately from the reaction mixture recovered from (12), conveyed to the distillation zone (22).

In a *"cascade"* arrangement of reactors, however, the reaction mixture is conveyed from one reactor to a second and optionally third ... etc. reactor before being separated into components. The principle is illustrated, for example, in the enclosed copy from ***Ullmann's*** *Encyclopedia of Industrial Chemistry*⁴⁾, page 96, Figure (G), where a liquid reaction mixture (a)⁵⁾ is charged at the top of the upmost first reactor vessel. The reaction mixture is then withdrawn at the bottom of the first vessel and conveyed to the top of the second reactor vessel. Discharge of the reaction mixture from a reactor vessel is, in each case, from the bottom, and charge of the reaction mixture to a subsequent reactor vessel is, in each case, to the top of the reactor. The Examiner will note that the reaction mixture passes through all of the reaction vessels of the reactor cascade

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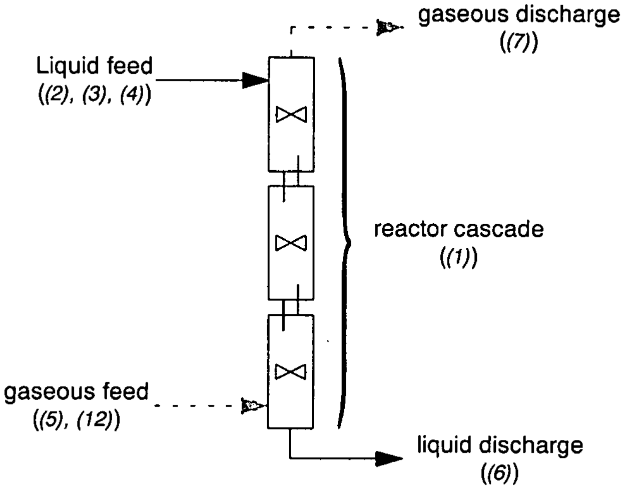
3) Note col. 3, indicated line 61, of ***US 4,661,624***.

4) Vol. B4, VCH Verlagsgesellschaft mbH, Weinheim 1992

5) (c) in Figure (G) signifies the inlet and outlet for a coolant supplied to the wall of the respective reactor vessel.

before it is finally discharged and, for example, conveyed to equipment suitable to separate the mixture into its components. Figure (G) of ***Ullmann's*** encyclopedia is illustrative of a cascade arrangement wherein the reactants are conveyed co-currently. A counter-current cascade reactor arrangement which is equally suitable for applicants' process[6] is illustrated in applicants' figure and schematically reproduced below:



In a counter-current reactor cascade the gaseous reactant[7] is introduced into the first reactor of the reactor cascade and is withdrawn from the last reactor of the reactor cascade[8], whereas the liquid reactants[9] are fed to the last reactor of the reactor cascade and are withdrawn from the first reactor of the cascade[10]. It is a characteristic of the reactor cascade arrangement, in co-current as well as in counter-current operation, that each of the reactants has to pass through all of the reactors of the cascade before being discharged.

In accordance with ***Chang et al.***'s disclosure and illustration, a stream of liquid reactants (*stream (14)*) is introduced into reaction zone (12) and a liquid product stream (28) is discharged from (12) and is conveyed to the distillation zone (22). Correspondingly, a stream of liquid reactants (*stream (20)*) is introduced into reaction zone (18) and a liquid product stream (30) is discharged from (18) and is conveyed to the distillation zone (22). Accordingly, there is no flow of liquid reactants and/or product from one the reaction zones to the other reaction zone before the product stream is discharged. The arrangement of ***Chang et al.***'s reactors (12) and (18) is therefore clearly not a *"cascade"* arrangement and requirement (1) of applicants' process is a first feature which distinguishes applicants' process from the process of ***Chang et al.***

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[6] Note page 11, indicated lines 5 to 10, of the application.

[7] Streams (5) and (12) in applicants' figure, compare page 23, indicated lines 6 to 14, of the application.

[8] Stream (7) in applicants' figure, compare page 23, indicated lines 6 to 14, of the application.

[9] Streams (2), (3) and (4) in applicants' figure, compare page 23, indicated lines 6 to 14, of the application.

[10] Stream (6) in applicants' figure, compare page 23, indicated lines 6 to 14, of the application.

In accordance with requirement (4) of applicants' invention, the reaction is carried out in the presence of from 0.05 to 0.5% by weight, based on the weight of the liquid feed, of a catalyst. In contrast thereto, ***Chang et al.*** teach that the catalyst is employed in amounts of from 2.0 to 6.0 mole-%, corresponding to 1.67 to 12.79% by weight11). The Examiner takes the position that a person of ordinary skill in the art would have been motivated by the statement of ***Chang et al.*** in col. 1, indicated lines 40 to 43, of ***US 4,661,624*** to employ the catalyst in lower concentrations. Applicants respectfully disagree. The referenced statement reads:

> *Also, when using the low catalyst concentrations of the prior art, the reaction will be slow and require long residence times, and thus substantial equipment costs.*

The distinct disadvantages which result, according to ***Chang et al.***'s statement, when the catalysts are employed in low concentrations clearly cannot be considered to motivate a person of ordinary skill to reduce the concentration of the catalyst in the process of ***Chang et al.*** However, the teaching or suggestion to make the necessary modification must be found in the prior art and cannot be based on applicants' disclosure and the same applies where the reasonable expectation of success is concerned12).

The Examiner has, in his argument, not particularly addressed requirements (2), (3) and (5) of applicants' process and merely notes in passing that applicants' process is conducted at *"superatmospheric pressure"* and that the process of ***Chang et al.*** is conducted at *"relatively high CO partial pressures"*. Applicants' respective requirements and the corresponding disclosure of ***Chang et al.*** are, therefore, merely briefly compared in form of a table:

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11) Col. 2, at indicated line 53, of ***US 4,661,624***.

12) In re Vaeck, 947 F.2d 488, 20 USPQ2d 1438 (CAFC 1991)

Requirement		Applicants' Claim 1	*Chang et al.* (US 4,661,624)	
(2)	reaction temperature	$T = 80 - 120^{\circ}C$	$T = 70 - 130^{\circ}C$	col. 2, indicated line 51
(3)	CO pressure	$p_{CO} = 90 - 180$ bar	$p_{CO} = 70 - 1000$ pisa corresponding to **$p_{CO} = 4.8 - 68.9$ bar**	col. 2, indicated line 52
(5)	molar ratio of CO:MeOH	0.5:1 - 3:1	0.1:1 - 0.6:1	col. 5, indicated lines 29 and 30

It is immediately apparent from the foregoing compilation that applicants' process differs significantly from the disclosure of ***Chang et al.*** in the CO partial pressure (3) and in the catalyst concentration (4).

With regard to requirements (7) to (11) of applicants' process the Examiner refers to ***Chang et al.***'s disclosure in col. 4, indicated lines 10 to 20, of ***US 4,661,624***, which provides that the liquid phase recovered at the bottom of the distillation zone (22) of ***Chang et al.***'s process is recycled to reactors (12) and (18), and asserts -referring to Claim 4- that catalyst desalting is integral in element (22) of ***Chang et al.*** Claim 4 of ***Chang et al.*** reads

> *A process as claimed in claim 1 wherein the CO containing stream is a purified coal or heavy residual oil gasification stream.*

and applicants find not support for the Examiner's assertion in that claim. Moreover, applicants' find nothing in the teaching of ***Chang et al.*** which suggests or implies that the distillation zone (22) has "desalting" capacity. Also, the statements of ***Chang et al.*** in col. 5, indicated lines 35 to 48, of ***US 4,661,624*** which were cited by the Examiner would appear to indicate that no desalting is deemed necessary in the process of ***Chang et al.*** because precipitation is avoided.

In accordance with the disclosure and illustration of ***Chang et al.***, essentially pure methanol is recovered from the bottom of the distillation zone (22) though line (26)[13], and a stream (34) of the recycled methanol which contains catalyst is removed, while -downstream from line (34)- fresh catalyst is introduced into the methanol stream through line (36) to maintain the desired catalyst activity. The fate of the stream (34) is not further addressed by ***Chang et al.*** As such, the disclosure of ***Chang et al.*** is silent with regard to requirements (8), (10) and (11) of applicants' process.

As explained in the foregoing, applicants' process is therefore distinguished over the teaching of ***Chang et al.*** because of applicants' requirements

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[13] Col. 5, indicated lines 3 et seq., of ***US 4,661,624***.

1) that the reaction is carried out in a cascade comprising at least two reactor elements;
3) that the reaction is carried out under a carbon monoxide pressure of from 90 to 180 bar;
4) that the reaction is carried out in the presence of from 0.05 to 0.5% by weight, based on the weight of the liquid feed, of an alkali metal alkoxide or alkaline earth metal alkoxide as catalyst;
8) that from 20 to 80% of the liquid phase which is recovered from the distilling apparatus is split off (part TR);
9) that the part TR is recirculated to the reactor cascade;
10) that the remaining part (*from 20 to 80%*) of the liquid phase which is recovered from the distilling apparatus (part TA) is discharged; and
11) that residual catalyst and catalyst degradation products are removed solids-free from the discharged part TA in a desalting apparatus and the remaining methanol is returned directly or indirectly to the reactor.

The disclosure of ***Chang et al.*** fails to suggest or imply either one of the foregoing elements of applicants' process. As previously submitted by applicants, to establish a prima facie case of obviousness, three basic criteria must be met:

(1) there must be some suggestion or motivation, either in the reference itself or in the knowledge generally available to one of ordinary skill in the art, to modify the reference.
(2) there must be a reasonable expectation of success.
(3) the prior art reference must teach or suggest all the claim limitations.

Moreover, the teaching or suggestion to make the claimed combination as well as the reasonable expectation of success must be found in the prior art and cannot be based on the applicant's disclosure14).

None of those three basic criteria is met where the teaching of ***Chang et al.*** and applicants' process are concerned. There is nothing in the teaching of ***Chang et al.*** -or in the knowledge generally available to one of ordinary skill in the art- which would motivate such a

---

14) See ftn. (12), page 6 of this paper.

person of ordinary skill to change and modify ***Chang et al.***'s process as is necessary to arrive at applicants' process. As such, the teaching of ***Chang et al.*** cannot be considered to teach or suggest all of the limitations which characterize applicants' process. At best, the teaching of ***Chang et al.*** invites a person of ordinary skill to try and change one or more of the process parameters set by ***Chang et al.*** in an attempt to improve upon the results achieved by ***Chang et al.*** In such a "trial and error" approach to modifying the prior art teaching there is, however, no reasonable expectation that one particular modification or combination of modifications will be successful. It is well settled that "obvious to try" is insufficient in a determination under Section 103(a)15).

For the foregoing reasons, the Examiner's position that the teaching of ***Chang et al.*** renders the subject matter of applicants' Claims 1 to 4 obvious within the meaning of Section 103(a) is deemed to be in error. Withdrawal of the respective rejection is respectfully solicited.

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Encl.: ***Ullmann's*** Encyclopedia of Industrial Chemistry, 5th Ed., Vol. B4, page 96

HBK/BAS

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15) In re O'Farrell, 853 F.2d 894, 903, 7 USPQ2d 1673, 1681 (CAFC 1988); see also In re Eli Lilly & Co., 902 F.2d 943, 14 USPQ2d 1741 (CAFC 1990) and In re Ball Corp., 925 F.2d 1480, 18 USPQ2d 1491 (CAFC 1991) (unpublished)

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Fifth, Completely Revised Edition

Volume B 4
Principles of Chemical Reaction Engineering and Plant Design

Editors: Barbara Elvers, Stephen Hawkins, Gail Schulz

VCH

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Principles of Chemical Reaction Engineering

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In addition to the standard symbols defined in the front matter of this volume, the following symbols are used:

a	specific contact area between reaction and nonreaction phase, m^2/m^3 dispersion
a_A	chemical activity of component A, mol/m^3
a_g	$= \frac{\lambda_g}{\varrho_g C_{p,g}}$; gas thermal diffusivity, m^2/s
a_{gl}	specific gas–liquid interface area per unit of dispersion volume, m^{-1}
a_{ls}	specific catalyst particle external surface area per unit of dispersion volume, m^{-1}
A_p	pellet external surface area, m^2
Al	$= \frac{(1-\varepsilon) k_{1,A}}{a \mathbb{D}_A}$; Hinterland ratio, ratio between total liquid-phase volume and liquid-phase stagnant film volume
An	$= \frac{E_a}{\Delta T_{ad} R} Le^{2/3}$; modified Arrhenius number
b_A	adsorption constant of component A for slow surface reaction and relatively fast ad- and desorption, Pa^{-1}
$b_{A,\infty}$	pre-exponential factor of b_A, Pa^{-1}
Bi_A	$= \frac{k_{g,A} r_p}{\mathbb{D}_{i,A}}$; Biot number of component A used in the shrinking core model
cosh(u)	hyperbolic cosine function: $\cosh(u) = \frac{e^u + e^{-u}}{2}$
C	total molar concentration of the reaction mixture, mol/m^3
C_A	concentration of component A, mol/m^3
$\overline{C_A}$	concentration of component A in well-mixed fluid bulk, mol/m^3
$C_{A,c}$	concentration of component A at core boundary in shrinking core model, mol/m^3
$\overline{C_{A,g}}$	concentration of component A in well-mixed gas phase bulk, mol/m^3
$C_{A,i}$	concentration of component A at interface, mol/m^3
$C_{A,in}$	concentration of component A in reactor inlet stream, mol/m^3
$C_{A,i,g}$	concentration of component A at gas–liquid interface on gas-phase side, mol/m^3
$C_{A,i,l}$	concentration of component A at gas–liquid interface on liquid-phase side, mol/m^3
$C_{A,i,s}$	concentration of component A at catalyst pellet external surface, mol/m^3
$\overline{C_{A,l}}$	concentration of component A in well-mixed bulk of liquid phase, mol/m^3
$C_{A,p}$	concentration of component A inside catalyst pellet with negligible intraparticle concentration gradient, mol/m^3
C_{A0}	liquid inlet concentration of component A for bubble column, mol/m^3
$C_{A,0}$	concentration of component A for zero conversion, mol/m^3
$C_A\|_t$	concentration of component A at time t, mol/m^3
$C_A\|_{t=0}$	concentration of component A at $t = 0$, mol/m^3
$C_A\|_z$	concentration of component A at location z in plug-flow reactor, mol/m^3
$C_A\|_{z=0}$	concentration of component A at $z = 0$ or at inlet of plug-flow reactor, mol/m^3
$C_{A,BR}(t)$	concentration of component A in batch reactor after reaction time t, mol/m^3
C_I	initiator concentration, mol/m^3
C_M	monomer concentration, mol/m^3
C_{MO}	concentration of metal oxide in virgin ore for shrinking core model, mol/m^3
$C_{p,g}$	specific heat of the gas, $J kg^{-1} K^{-1}$
$C_{P^\cdot}$	concentration of growing, free-radical polymer chains, mol/m^3
$C_{R^\cdot}$	free-radical concentration, mol/m^3
C_T	concentration of tracer material, mol/m^3
d_b	gas bubble diameter, m
d_S	$= \frac{\sum d^3}{\sum d^2}$; Sauter mean bubble diameter, m
Da	$= \frac{kt}{C_{A,0}}$ (BR), $\frac{k\tau}{C_A\|_{z=0}}$ (PFR), $\frac{k\tau}{C_{A,in}}$ (CISTR and CSSTR); Damköhler number for zeroth-order kinetics
$\mathbb{D}_A$	molecular diffusion coefficient of component A, m^2/s
$\mathbb{D}_{A,g}$	diffusion coefficient of component A in gas phase, m^2/s
$\mathbb{D}_{fl}$	diffusion coefficient in bulk of fluid, m^2/s
$\mathbb{D}_i$	effective diffusion coefficient inside porous pellet, m^2/s
$\mathbb{D}_{i,A}$	effective diffusion coefficient inside porous pellet of component A, m^2/s
$\mathbb{D}^h_{i,A}$	effective diffusion coefficient in ring-shaped or cylindrical porous catalyst pellet in longitudinal direction, m^2/s
$\mathbb{D}^*_{i,A}$	effective diffusion coefficient, modified for pellet anisotropy, m^2/s
$\mathbb{D}^r_{i,A}$	effective diffusion coefficient in ring-shaped or cylindrical porous catalyst pellet in radial direction, m^2/s
$\mathbb{D}_{Kn}$	Knudsen diffusion coefficent, m^2/s
erf(u)	error function: $\mathrm{erf}(u) = \frac{2}{\sqrt{\pi}} \int_0^u \exp(-x^2)\, \partial x$
E_a	activation energy, J/mol

E_A	$= \frac{\overline{J_A} \text{ with reaction}}{\overline{J_A} \text{ without reaction}}$; enhancement factor of component A
E_{A0}	$= \frac{\overline{J_A}}{k_{l,A} C_{A,i}}$; modified enhancement factor used, e.g., in Van Krevelen–Hoftijzer approximation
$E_{A\infty}$	$= \left(1 + \frac{1}{\nu} \frac{\mathbb{D}_B}{\mathbb{D}_A} \frac{\overline{C_B}}{C_{A,i}}\right) \left(\frac{\mathbb{D}_B}{\mathbb{D}_A}\right)^n$; enhancement factor for case of instantaneous reaction
E_+	activation energy for reaction to right-hand side, J/mol
E_-	activation energy for reaction to left-hand side, J/mol
$E(t)$	residence-time frequency distribution as function of t, s^{-1}
$E(\theta)$	residence-time frequency distribution as function of θ
f	initiator efficiency; for $f = 1$, all initiator decomposes to active radicals; for $f = 0$, no active radicals are formed at all
$F(t)$	$= \int_0^t E(u)\,\partial u$; cumulative distribution of residence times
h	longitudinal coordinate, m
H	(1) parallelepiped height, m (2) height of Raschig ring, m
Ha	Hatta number, measure of square root of reaction rate divided by rate of mass transfer of component A; exact definition depends on kinetic expression
Ha_i	modified Hatta number for reactions between reactants from a gas and a liquid, based on liquid-phase reactant interface concentration $C_{B,i}$ (usual Hatta number is based on bulk concentration $\overline{C_B}$)
$(-\Delta H_{ads})$	adsorption enthalpy of component A on chemically active catalyst surface; since adsorption is an exothermic process, $(-\Delta H_{ads}) > 0$, J/mol
$(-\Delta H_r)$	reaction enthalpy; for exothermic reactions, $(-\Delta H_r) > 0$; for endothermic reactions, $(-\Delta H_r) < 0$, J/mol
$I_0(u)$	zeroth-order modified Bessel function of the first kind: $I_0(u) = \sum_{i=0}^{\infty} \left(\frac{u^i}{2^i\, i!}\right)^2$
$I_1(u)$	first-order modified Bessel function of the first kind: $I_1(u) = \frac{u}{2} \sum_{i=0}^{\infty} \frac{(u/2)^{2i}}{i!\,(i+1)!}$
J_A	molar flux of component A through interface, $mol\, m^{-2} s^{-1}$
$\overline{J_A}$	average molar flux of component A for contact time τ of a stagnant fluid element in penetration theory, $mol\, m^{-2} s^{-1}$
$J_{A,p}$	molar flux of component A at pellet external surface, $mol\, m^{-2} s^{-1}$
$(J_A)_{phys}$	molar flux of component A through interface if no chemical reaction occurs, $mol\, m^{-2} s^{-1}$
$(J_A)_x$	molar flux of component A at location x, $mol\, m^{-2} s^{-1}$
$J_A(r)$	molar flux of component A at location r, $mol\, m^{-2} s^{-1}$
k	reaction rate constant; dimension depends on kinetic expression
k_g	gas-phase mass-transfer coefficient for transfer from a gas to a liquid, m/s
$k_{g,A}$	mass-transfer coefficient of component A in a gas, m/s
k_l	liquid-phase mass-transfer coefficient for transfer from a gas to a liquid, m/s
k_{ls}	liquid-phase mass-transfer coefficient for transfer from a liquid to a solid, m/s
$k_{l,A}$	mass-transfer coefficient of component A in a liquid, m/s
$k_{n,m}$	reaction rate constant for bimolecular reactions with (n,m)-kinetics, $m^{3(n+m-1)}\ (mol\, A)^{-(n-1)}\ (mol\, B)^{-m}$, s^{-1}
k_p	reaction rate constant based on pellet volume; dimension depends on kinetic expression
$k_{p,0}$	pre-exponential factor for rate constant k_p (based on catalyst pellet volume)
$k_p(T_p)$	reaction rate constant based on pellet volume for first-order kinetics at pellet temperature T_p, s^{-1}
k_r	reaction rate constant; dimension depends on kinetic expression
k_r''	surface reaction rate constant, m/s
k_+	reaction rate constant of reaction to right-hand side; dimension depends on kinetic expression
k_-	reaction rate constant of reaction to left-hand side; dimension depends on kinetic expression
k_∞	reaction rate constant pre-exponential factor; reaction rate constant for infinite temperature; dimension depends on kinetic expression
K	$= \frac{C_P}{C_A}, \frac{C_P C_Q}{C_A C_B}$; chemical equilibrium constant
K_A	adsorption constant of component A, m^3/mol
K_S	saturation constant in biochemical reactions, kg/m^3
$K_0(u)$	zeroth-order modified Bessel function of the second kind: $K_0(u) = -\ln(u/2)\, I_0(u) + \sum_{i=1}^{\infty} \left\{ psi(i+1) \left(\frac{u^i}{2^i\, i!}\right)^2 \right\}$
$K_1(u)$	first-order modified Bessel function of the second kind: $K_1(u) = \ln(u/2)\, I_1(u) + \frac{u}{2} \sum_{i=1}^{\infty} \left\{ \frac{psi(i+1) + psi(i+2)}{2} \frac{(u/2)^{2i}}{i!\,(i+1)!} \right\}$
L	(1) length of plug-flow reactor, m (2) parallelepiped length, m
Le	$= \frac{a_g}{\mathbb{D}_{A,g}}$; gas Lewis number for component A
m	(1) (apparent) reaction order of component B (2) $= \frac{C_{A,i,l}}{C_{A,i,g}}$; dimensionless distribution coefficient or equilibrium constant of component A between gas and liquid phases
mc_k	$\int_0^\infty (\theta - 1)^k E(\theta)\,\partial\theta$; k-th central moment of dimensionless frequency distribution
mo_k	$\int_0^\infty \theta^k E(\theta)\,\partial\theta$; k-th moment around origin of dimensionless frequency distribution
M_A	molar mass of component A, kg/mol
M_{tot}	total reaction mixture mass in closed system, kg
MC_k	$\int_0^\infty (t - \bar{t})^k E(t)\,\partial t$; k-th central moment of frequency distribution, s^k
MO_k	$\int_0^\infty t^k E(t)\,\partial t$; k-th moment around origin of frequency distribution, s^k

n	(apparent) reaction order of component A
n_{A}	number of moles of component A, mol
$n_{\mathrm{A},0}$	number of moles of component A for zero conversion, mol
$n_{\mathrm{T},0}$	number of moles of tracer injected in a pulse at $t = 0$, mol
psi (u)	psi function; digamma function: $\mathrm{psi}(u) = -\gamma + \sum_{i=1}^{u-1} \frac{1}{i} \quad u \in \mathbb{Z}^+$
P_{A}	partial pressure of component A, Pa
r	radial coordinate, m
r^*	radial coordinate in ring-shaped or cylindrical porous catalyst pellet, modified for pellet anisotropy, m
r_{c}	core radius in shrinking core model, m
r_{p}	(1) pellet radius in shrinking core model, m (2) catalyst pellet radius, m
R	ideal gas constant ($8.31434\ \mathrm{J\,mol^{-1}\,K^{-1}}$)
R_{A}	chemical production rate of component A (if A is consumed, $R_{\mathrm{A}} < 0$; if A is produced, $R_{\mathrm{A}} > 0$), $\mathrm{mol\,m^{-3}\,s^{-1}}$
$\lvert R_{\mathrm{A}} \rvert$	chemical conversion rate of component A (positive if reaction proceeds in direction of arrow), $\mathrm{mol\,m^{-3}\,s^{-1}}$
$\lvert R_{\mathrm{A}} \rvert_{\mathrm{p}}$	conversion rate of component A based on catalyst pellet volume, $\mathrm{mol\,(m^3\,pellet)^{-1}\,s^{-1}}$
$\lvert R_{\mathrm{A}} \rvert_{\mathrm{P}}$	conversion rate of component A toward desired product P, $\mathrm{mol\,m^{-3}\,s^{-1}}$
$\lvert \mathbb{R}_{\mathrm{A}} \rvert_{\mathrm{X}}$	conversion rate of component A toward undesired byproduct X, $\mathrm{mol\,(m^{-3}\,s^{-1})}$
R_{i}	inner radius of Raschig ring, m
R_{i}^*	Raschig ring inner radius modified for pellet anisotropy, m
R_{u}	outer radius of Raschig ring, m
R_{u}^*	Raschig ring outer radius modified for pellet anisotropy, m
$\lvert \mathbb{R}_{\mathrm{A}} \rvert$	chemical reaction rate of component A; rate in absence of mass- or heat-transfer limitation, $\mathrm{mol\,m^{-3}\,s^{-1}}$
$\overline{\lvert \mathbb{R}_{\mathrm{A}} \rvert}$	average reaction rate for two nonmixed fluid microelements, $\mathrm{mol\,m^{-3}\,s^{-1}}$
$\lvert \mathbb{R}''_{\mathrm{A}} \rvert$	reaction rate based on pellet external surface area, $\mathrm{mol\,(m^2}$ external surface area$\mathrm{)^{-1}\,s^{-1}}$
$\lvert \mathbb{R}_{\mathrm{A}} \rvert_{\mathrm{mixed}}$	reaction rate for two mixed fluid microelements, $\mathrm{mol\,m^{-3}\,s^{-1}}$
$\lvert \mathbb{R}_{\mathrm{A}} \rvert_{\mathrm{p}}$	reaction rate based on pellet volume, $\mathrm{mol\,m^{-3}\,s^{-1}}$
$\lvert \mathbb{R}''_{\mathrm{A}} \rvert_{\mathrm{P}}$	rate of reaction yielding desired product P based on pellet external surface area, $\mathrm{mol\,(m^2}$ external surface area$\mathrm{)^{-1}\,s^{-1}}$
$\lvert \mathbb{R}_{\mathrm{A}} \rvert_{\mathrm{r}}$	chemical reaction rate based on reaction mixture or reactor volume, $\mathrm{mol\,m^{-3}\,s^{-1}}$
$\lvert \mathbb{R}''_{\mathrm{A}} \rvert_{\mathrm{X}}$	reaction rate of reaction yielding undesired byproduct X based on the pellet external surface area, $\mathrm{mol\,(m^2}$ external surface area$\mathrm{)^{-1}\,s^{-1}}$
sinh(u)	hyperbolic sine function: $\sinh(u) = \frac{e^u - e^{-u}}{2}$
S	cross-sectional area of tube or plug-flow reactor, $\mathrm{m^2}$
s	fraction of stagnant fluid elements replaced per unit of time in Danckwerts penetration or surface renewal model, $\mathrm{s^{-1}}$
tanh(u)	hyperbolic tangent function: $\tanh(u) = \frac{e^{2u} - 1}{e^{2u} + 1}$
T	(1) reaction mixture temperature, K (2) temperature inside a catalyst pellet, K
$\bar{t}$	reactor average residence time, s
$t_{1\%}$	time lapse needed for polymer chain radical concentration to reach 99 % of steady-state value, s
ΔT_{ad}	$= \frac{\overline{C_{\mathrm{A,g}}}\,(-\Delta H_{\mathrm{r}})}{\varrho_{\mathrm{g}}\,C_{\mathrm{p,g}}}$; adiabatic temperature rise for monomolecular gas-phase reaction, K
$\overline{T_{\mathrm{g}}}$	temperature of gas bulk surrounding catalyst pellet, K
T_{i}	temperature at external surface of catalyst pellet, K
T_{max}	temperature at which the maximum rate is found for exothermic equilibrium reaction with fixed concentration in the reaction mixture, K
T_{p}	catalyst pellet temperature for negligible intraparticle temperature gradients, K
$T_{\mathrm{p,max}}$	$= \overline{T_{\mathrm{g}}} + \Delta T_{\mathrm{ad}}\, Le^{2/3}$; maximum pellet temperature for heterogeneously catalyzed gas-phase reaction, K
u	superficial velocity; velocity based on empty tube cross-sectional area, m/s
u_{b}	gas bubble rise velocity in liquid, m/s
u_x	fluid velocity in x-direction, m/s
u_y	fluid velocity in y-direction, m/s
u_z	fluid velocity in z-direction, m/s
U(u)	unit step function or Heaviside step function: $\mathrm{U}(u) = \begin{cases} 0 & u < 0 \\ 1 & u \geq 0 \end{cases}$
V	total reaction mixture volume, $\mathrm{m^3}$
V_{p}	pellet volume, $\mathrm{m^3}$
W	parallelepiped width, m
x	coordinate perpendicular to interface through which mass transfer occurs, m
z	axial coordinate in plug-flow reactor, m
α_{g}	heat-transfer coefficient between catalyst pellet and surrounding gas, $\mathrm{W\,m^{-2}\,K^{-1}}$
γ	$= \lim_{n \to \infty} \left(\sum_{i=1}^{n} \frac{1}{i} - \ln(n) \right) = 0.5772156649$; Eulers constant
γ_{A}	activity or fugacity coefficient of component A
γ_{B}	$= \upsilon_{\mathrm{B}} \frac{\mathbb{D}_{\mathrm{i,A}}}{\mathbb{D}_{\mathrm{i,B}}} \frac{C_{\mathrm{A,i}}}{C_{\mathrm{B,i}}}$; excess amount of component B for bimolecular reactions
γ_{p}	pore tortuosity factor for porous pellet
δ	film thickness in stagnant film model, m
δ_{A}	film thickness for component A in stagnant film model, m
δ_{r}	thickness of reaction zone in stagnant film with thickness $\delta > \delta_{\mathrm{r}}$, m
$\delta(u)$	Dirac delta function: $\delta(u) = \lim_{\varepsilon \to 0} \begin{cases} 0 & u < -\varepsilon, \quad u > \varepsilon \\ \frac{1}{2\varepsilon} & -\varepsilon \leq u \leq \varepsilon \end{cases}$
Δ_{FK}	error introduced on using Frank-Kamenetzki approximation
$\Delta_{\mathrm{FK,max}}$	maximum value of Δ_{FK} (i.e., for $C_{\mathrm{A}} = 0$ or low effectiveness factors)
ε	(1) porosity of packed bed of catalyst pellets; volume fraction void (2) volume fraction of gas in gas-in-liquid dispersion
ε_{p}	porosity of porous particle; fraction void volume
ζ_{A}	relative degree of conversion of component A
$\zeta_{\mathrm{A,BR}}$	relative degree of conversion of component A in batch reactor
$\zeta_{\mathrm{A,CISTR}}$	relative degree of conversion of component A in CISTR

$\zeta_{A,\mathrm{CSSTR}}$	relative degree of conversion of component A in CSSTR
$\zeta_{A,\mathrm{equi}}$	relative degree of conversion of component A at equilibrium conditions
$\zeta_{A,\mathrm{PFR}}$	relative degree of conversion of component A in plug-flow reactor
ζ_{MO}	relative degree of conversion of metal oxide in shrinking core model
$\zeta_{\mathrm{M,de}}$	maximum relative degree of conversion of monomer obtained for dead-end polymerization
η	effectiveness factor; amount of key reactant converted in porous catalyst pellet, divided by amount that could be converted if no retardation by diffusion were to occur
$\eta^{\ominus}$	$= \frac{\overline{J_A} a V}{k C_{A,i} (1-\varepsilon) V}$; degree of reactor utilization
η_P	$= \sigma_P \zeta_A$; yield of product P; ratio of amount of P formed to amount that could have been formed if all of key reactant A were converted solely to P
θ	$= \frac{t}{\tau}$; dimensionless residence time in reactor
θ_g	$= \frac{\overline{T_g}}{\Delta T_{ad}} Le^{2/3}$; dimensionless gas temperature
θ_p	$= \frac{T_p}{\Delta T_{ad}} Le^{2/3}$; dimensionless pellet temperature
ι	$= \frac{R_i}{R_u}$; dimensionless inner radius of Raschig ring
κ_A	mole fraction of component A
$\overline{\kappa_A}$	mole fraction of component A in bulk of fluid
$\kappa_{A,i}$	mole fraction of component A at interface
λ	$= \frac{H}{R_u}$; dimensionless height of Raschig ring
λ_g	gas thermal conductivity, $\mathrm{Wm^{-1}K^{-1}}$
λ_p	catalyst pellet thermal conductivity, $\mathrm{Wm^{-1}K^{-1}}$
μ	$= \frac{(-\Delta H_r) \mathbb{D}_{i,A} C_{A,i}}{\lambda_p T_i}$; measure of maximum relative temperature increase inside pellet
ν	stoichiometric coefficient
ν'	kg enzymes or cells per kg substrate in biochemical reactions
ν_B	stoichiometric coefficient of component B
ξ_0	$= \frac{k_g A_p}{k_{p,0} V_p}$; mass-transfer rate to catalyst pellet over reaction rate at infinitely high temperature
$\xi_{0,\mathrm{bo}}$	value of ξ_0 for which pellet blowout occurs
$\xi_{0,\mathrm{ra}}$	value of ξ_0 for which pellet runaway occurs
$\xi(T_p)$	$= \frac{k_g A_p}{k_p(T_p) V_p}$; measure of mass-transfer rate to catalyst pellet divided by reaction rate
ϱ	(1) density of reaction mixture, $\mathrm{kg/m^3}$ (2) $= \frac{r_c}{r_p}$; dimensionless core radius in shrinking core model
ϱ_g	gas density, $\mathrm{kg/m^3}$
ϱ_0	density of reaction mixture for zero conversion, $\mathrm{kg/m^3}$
σ_P	selectivity toward product P; ratio of amount of P formed over amount that could have been formed if all of key reactant A had been converted to P
$\sigma_{P,Ha<0.2}$	selectivity toward desired product P for parallel reactions in slow reaction regime $Ha < 0.2$
$\sigma_{P,Ha>2}$	selectivity toward desired product P for parallel reactions in fast reaction regime $Ha > 2$
σ_t	$= \sqrt{\sigma_t^2}$; standard deviation of frequency distribution, s
σ_t^2	$= \mathrm{MC}_2$; variance of frequency distribution, $\mathrm{s^2}$
σ_θ^2	$= \mathrm{mc}_2$; variance of dimensionless frequency distribution
τ	(1) reactor holding time, s (2) contact time between small stagnant fluid element and interface in Higbie penetration model, s
τ_D	characteristic time for shrinking core in regime of slow diffusion through converted shell, s
τ_L	$= \frac{(1-\varepsilon)V}{\varphi_{V\,l}}$; average residence time in liquid phase in bubble column, s
τ_M	characteristic time for shrinking core in regime of slow external mass transfer, s
τ_R	characteristic time for shrinking core in regime of slow reaction kinetics, s
φ	$\equiv \frac{1}{\eta}$ for low η values thus high φ values; generalized Thiele modulus
φ_A	$= \frac{V_p}{A_p} \sqrt{\frac{k_p}{\mathbb{D}_{i,A}}}$; Aris' shape-generalized Thiele modulus for first-order kinetics
φ_m	mass flow rate, kg/s
φ_T	$= r_p \sqrt{\frac{k_p}{\mathbb{D}_{i,A}}}$; simple Thiele modulus for first-order kinetics; not generalized for arbitrary catalyst geometries
φ_v	volumetric flow rate, $\mathrm{m^3/s}$
$\varphi_{v,0}$	volumetric flow rate of liquid feed to bubble column, $\mathrm{m^3/s}$
$\varphi_{v,1}$	volumetric flow rate of liquid stream out of bubble column, $\mathrm{m^3/s}$
χ	$= \frac{E_a}{RT_i} \cdot \frac{(-\Delta H_r) \mathbb{D}_{i,A} C_{A,i}}{\lambda_p T_i}$; measure of maximum relative reaction rate increase inside pellet due to intraparticle temperature gradients
ψ''	heat flux from or to catalyst pellet, $\mathrm{W/m^2}$
ω_A	mass fraction of component A
$\omega_{A,0}$	mass fraction of component A for zero conversion

1. Introduction

A rational approach to all problems relating to a physical or chemical change of matter must be based on elementary physical conservation laws. They have been handled systematically in, for example, [1] and [2]. For the treatment of chemical engineering phenomena, the laws of conservation of matter and energy are of primary importance; they are the basis of the discussions below. Flow phenomena, governed by the principle of conservation of momentum, naturally are equally important for chemical engineering calculations (→ 5. Fluid Mechanics, **B 1**).

The second kind of information needed for a quantitative treatment of chemical engineering is

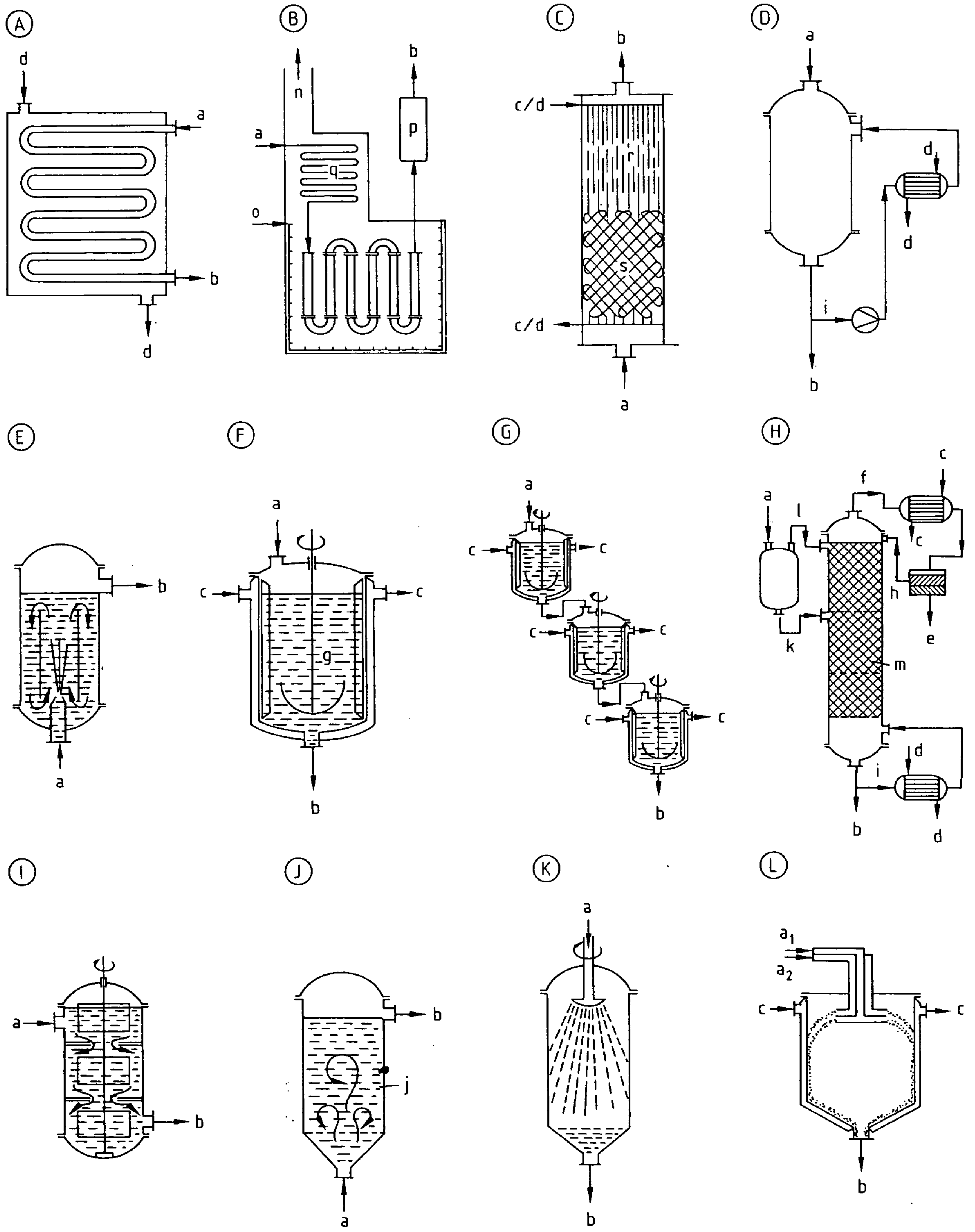
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Figure 3. Reactors for liquid-phase reactions

A) Tubular reactor; B) Reformer; C) Sulzer mixer – reactor; D) Reactor with external recirculation; E) Reactor with internal recirculation (draft tube); F) Stirred tank; G) Cascade of stirred tanks; H) Column reactor; I) Multichamber tank; J) Fluidized-bed reactor; K) Spray reactor; L) Falling-film reactor

a) Liquid reaction mixture; a_1, a_2) Liquid feed components; b) Liquid product; c) Coolant; d) Heating agent; e) Water; f) Organic phase and water; g) Baffle; h) Organic phase; i) Partial stream of product; j) Catalyst; k) Reaction mixture from preceding reaction stage; l) Water from preceding stage; m) Packing; n) Off-gas; o) Fuel gas for burners; p) Quench; q) Convection zone; r) Mixing element consisting of tubes carrying heat-transfer medium; s) Mixing elements rotated 90°